This Page Is Inserted by IFW Operations and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problems Mailbox.

THIS PAGE BLANK (USPTO)

(12) UK Patent Application (19) GB (11) 2 188 559 (13) A

(43) Application published 7 Oct 1987

- (21) Application No 8705753
- (22) Date of filing 11 Mar 1987
- (30) Priority data
 - (31) 3608635
- (32) 14 Mar 1986
- (33) DE
- (71) Applicant
 Drache Keramikfilter Produktions-GmbH,

(Incorporated in FR Germany),

Werner-von-Siemens-Strasse 9, 6252 Diez, Federal Republic of Germany

- (72) Inventors
 Helmut Schweers Dipl.-Ing,
 Günther Förster Dipl.-Ing,
 Frank Drache Dipl.-Betriebswirt
- (74) Agent and/or Address for Service
 F. J. Cleveland & Company, 40-43 Chancery Lane,
 London WC2A 1JQ

- (51) INTCL⁴ 801D 53/36
- (52) Domestic classification (Edition I) B1F D2D U1S 1851 B1F
- (56) Documents cited

GB 1461804 GB 1455433 GB 1406704 US 4581206 US 4425304 US 4278639

(58) Field of search

B1F

Selected US specifications from IPC sub-class B01D

(54) Exhaust gas reactor

(57) An exhaust gas reactor comprises at least first and second open-pore ceramic foam plates which are arranged in succession in the direction of flow through the reactor. The plates of ceramic foam have a catalytically active surface layer thereon. Filter plates of uncoated ceramic foam may additionally be provided upstream and downstream of the first-mentioned plates.

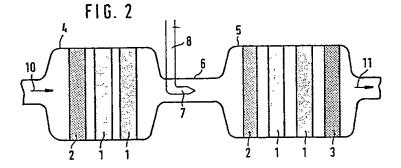
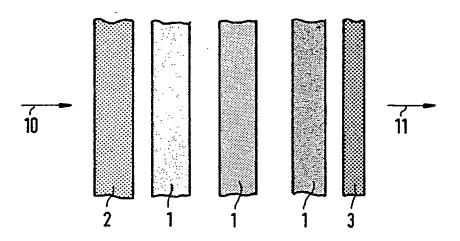
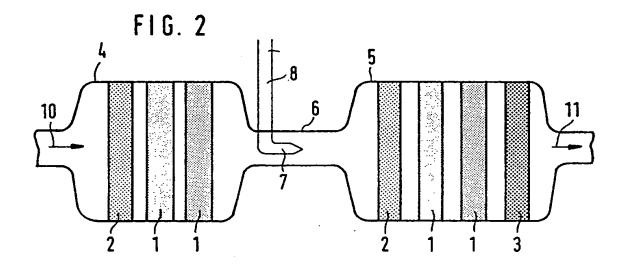


FIG. 1





5

SPECIFICATION

Exhaust gas reactor and process for the production thereof

The invention relates to an exhaust gas reactor, for example for use with an internal combustion engine, and a process for the production of such a reactor.

The term exhaust gas is used in this specification
10 in a broad sense and may therefore embrace exhaust
gas from an internal combustion engine as well as
exhaust or waste gas from other sources, such as a
paint spraying booth.

One form of an exhaust gas reactor comprises a
15 refractory carrier material in the form of a monolithic
block of open pore ceramic foam, which has thereon
a catalytically active surface layer.

Porous ceramic foam is produced by an open-pore plastics foam being filled with a slurry of ceramic

20 material, and then vaporising the plastics foam at elevated temperature. The porous ceramic foam produced in that way is also of an open-pore nature and is then provided with a catalytic layer which is matched to the respective chemical process

25 involved. Catalysts produced in that way from porous ceramic foam are subjected to further

porous ceramic foam are subjected to further processing to produce monolithic blocks of appropriate size, and used in that form.

According to the present invention, there is
provided an exhaust gas reactor including a
refractory carrier means of open-pore ceramic foam
comprising at least two plates which are disposed in
succession in the direction of gas flow through the
reactor, and a catalytically active surface layer on the
carrier means.

Tests have been carried out which show that an exhaust gas reactor which is constructed from a plurality of successively disposed plates is substantially more efficient in operation than an 40 exhaust gas reactor consisting of a monolithic block of appropriate size. Furthermore, an exhaust gas reactor comprising a plurality of plates is substantially easier to manufacture and it is also easier to provide it with a catalytically active surface 45 layer, than a large monolithic block. It is possible to arrive at a made-to-measure construction, depending on the particular composition and temperature of the waste or exhaust gases which are to be subjected to the post-combustion procedure,

50 by virtue of the plates being of suitable composition, configuration and dimensions. In addition, the reactor can be rapidly adapted to the respective operating conditions involved, particularly in the event of changes therein, by virtue of the insertion or replacement of plates. All in all, because of the large internal surface area of the ceramic foam, the

5 replacement of plates. All in all, because of the large internal surface area of the ceramic foam, the exhaust gas reactor can be small in volume, in relation to the respective gas through-put thereof. A compact exhaust gas reactor of that nature is

60 particularly important when, in connection with subsequent fitting to motor vehicles, a conventional silencer box or muffler is to be replaced by an exhaust gas reactor which must take account of the predetermined dimensions and conditions of

65 installation of the respective silencer box or muffler.

Besides the very large internal surface area of a ceramic foam, relative to its external volume, a critical consideration in regard to the level of efficiency of an exhaust gas reactor in accordance with the present invention is also the high level of reaction probability which is provided by the micro-turbulent post-flow of the individual rods or fibres of the ceramic foam.

In accordance with a further feature of the
75 invention, the porosity of the plates is between 10
and 60 ppi (pores per inch), and preferably between
10 and 30 ppi. The reference to pores per inch (ppi)
denotes the number of pores per inch as a unit of
length.

A further embodiment of a reactor according to the invention advantageously provides that plates of a different configuration or nature may be used. The different configuration or nature may be that the porosity of the plates is different, for example a plate with coarse pores is followed by one or more plates with finer pores. However the different configuration or nature may additionally or alternatively involve different catalytically active surface layers.

Furthermore and possibly in addition thereto the plates may be of different shapes and sizes in order for example to produce a given flow characteristic in aerodynamic respects.

A further feature of the invention may provide that

one or more uncoated plates is or are arranged 95 upstream and/or between and/or downstream of catalytically coated plates. In that connection the uncoated plates act in particular as a filter means. In that way, undesirable particles in the gas flowing into the reactor are filtered out of the gas flow prior to 100 passing into the part of the reactor in which reaction occurs, and reaction products are filtered out of the gas flow if further uncoated plates are arranged within the coated plates of the reactor or downstream thereof. The pore size of the uncoated 105 filter plates is to be matched to the size of the particles which are to be filtered out of the gas flow. If an uncoated plate with very fine pores is disposed at the downstream end of the reactor assembly, it is possible to ensure that any particles of the ceramic 110 carrier material which come loose do not pass into the atmosphere, together with constituents of the washcoat base layer and the catalyst material.

In one embodiment of a reactor, the individual plates thereof may bear against each other in a 115 sandwich-like construction. An advantageous feature of the invention provides however that the plates are arranged at a spacing from each other, in which respect the spacings between the plates may be the same or may vary. The spacings between the 120 plates prevent the gases from forming preferential through-flow paths, which would have the result that parts of the plates would not have gas flowing therethrough or would have less gas flowing therethrough, so that the level of efficiency would 125 drop, in relation to unit of surface area. If the spacing between plates is correctly chosen, turbulent effects occur between the plates, which ensure that the gas flow is balanced and equalised out. The spacings between the plates are also to be selected having 130 regard to the flow speeds through the reactor, the

dimensions of the plates and the properties of the respective gases involved. In that connection however the spacings should be kept as small as possible, having regard to the reactor being of a 5 compact design.

If, in accordance with a further advantageous feature of the invention, the plates are individually interchangeable, then a reactor of that nature is simpler and less expensive from the point of view of 10 maintenance than a reactor which uses a monolithic block. In that case individual plates can be replaced for repair or exchanged for other plates, if the operating conditions alter. To remove the dust which has been produced, the above-mentioned uncoated filter plates may be exchanged or only cleaned after they have been removed.

A particularly advantageous development which is also an aspect of independent significance provides that the catalyst material used comprises at least one 20 non-noble metal oxide.

The reaction temperature of such oxides is higher than that of noble metals which are conventionally used, for example platinum, and is between about 450 and 750°C. As a result, the level of effectiveness scarcely decreases, even under high loading and at high temperatures, so that such a reactor is particularly suitable not just for internal combustion engines of motor vehicles but also for stationary engines. The non-noble metal oxides are also resistant to lead poisoning phenomena and to other noxious substances such as chlorine, fluorine and sulphur, as occur in the uncleaned gases of stationary engines.

In a preferred feature of the invention, the
35 catalytically active surface layer of a non-noble metal oxide may be arranged on what is known as a washcoat base layer of aluminium oxide in its γ-modification form. Such a washcoat base layer is known and provides for a more rapid and better
40 reaction. In order to improve long-term stability, additives may be added to the base layer, for example compounds of elements from the rare earth group.

In order to provide a reducing reaction, a further 45 embodiment of the invention provides for using a catalyst material comprising at least one oxide of the following metals: iron (Fe₂O₃ or Fe₃O₄), cobalt (CoO or Co₂O₃ or Co₃O₄), nickel (NiO or Ni₂O₃), titanium (TiO₂), vanadium (V₂O₅), chromium (Cr₂O₃), 50 'molybdenum (MoO₃), tungsten (WO₃), and manganese (MnO or Mn₂O₃ or Mn₃O₄), or at least a mixture of at least two thereof.

To provide an oxidising reaction, the catalyst material of the reactor may comprise at least one 55 oxide of at least one of the following metals: copper (CuO), chromium (Cr_2O_3), molybdenum (MoO_3), tungsten (WO_3), manganese (MnO or Mn_2O_3 or Mn_3O_4), vanadium (V_2O_5), iron (Fe_2O_3) or at least one mixture of at least two thereof.

60 In a further advantageous embodiment of the invention, the reactor may be of a two-stage configuration, with a reducing reaction taking place in the first stage and an oxidising reaction taking place in the second stage. For example, in the first

65 stage of an exhaust gas reactor for internal

combustion engines, nitrogen oxides (NO_x) are reduced to harmless nitrogen at the catalyst material by means of the carbon monoxide (CO) in the exhaust gas. Then, the second stage involves

70 oxidation of the excess carbon monoxide to form carbon dioxide, with oxygen possibly also being added, in the form of air. In other areas of use of such a reactor, it is also possible to provide for the oxidation of other constituents, in particular organic 75 components, for example hydrocarbons. Such

75 components, for example hydrocarbons. Such constituents occur inter alia in the waste or exhaust gases from paint spraying booths. The two stages of the reactor each have at least one plate and may be arranged in separate housings or in a common

80 housing. At least one uncoated filter plate is desirably arranged upstream of the first stage, so such a filter can also retain soot particles which can then undergo post-combustion if the initial filter reaches a temperature above 600°C. The second 85 stage may be followed by an uncoated fine-pore

5 stage may be followed by an uncoated fine-pore filter plate, constituting a very fine filter which retains in particular pieces of ceramic carrier material which have come away.

In another aspect, the present invention also
provides a process for the production of an exhaust
gas reactor, wherein a non-noble metal oxide is
added to the slurry which, as referred to in the
opening part of this specification, is used to produce
the ceramic foam from open-pore plastics foam.

95 After calcination of the foam which is coated with the ceramic material, the non-noble metal oxide is then a constituent of the ceramic material and is thus also present at the surface thereof. Such a mixing effect may be used in particular when the oxide is

100 comparatively inexpensive.

An alternative option in regard to the process for producing the exhaust gas reactor according to the invention provides that the foam which has been coated with ceramic material and pre-dried but not yet subjected to the calcination operation may be coated, preferably by a dipping operation, with a particular suspension or slurry which contains the non-noble metal oxide and optionally further constituents which promote adhesion of the oxide as well as the catalytic function. Besides for example titanium dioxide, the suspension or slurry may additionally contain the material for the base layer (aluminium oxide).

Another form of procedure for applying the
catalyst material in the reactor according to the
invention provides that the foam which has been
coated with ceramic material and possibly the base
layer and which may or may not have been subjected
to the calcination operation is impregnated with a
solution of a metal salt of the non-noble metal to be
used, and then the metal is converted into the metal
oxide by a heat treatment such as calcination, and
fixed on the ceramic foam.

The reactor according to the invention is

125 inexpensive to manufacture, enjoys a wide range of
adaptability and has a high level of efficiency, even at
high temperatures, while being easy and simple to
maintain and being capable of adapting to altering
operating conditions.

130 Embodiments of a reactor according to the

invention and the process for the production thereof will now be described by way of example with reference to the accompanying drawing in which:

Figure 1 is a diagrammatic view in longitudinal section of a first embodiment of the reactor according to the invention, and

Figure 2 is a basic view in longitudinal section of a two-stage reactor as a second embodiment of the invention.

10 Referring to the diagrammatic view in Figure 1, an exhaust or waste gas reactor according to the invention comprises three plates 1 of open-pore ceramic foam with a catalytically active surface layer thereon, which are arranged in succession in the direction in which the gas flows through the reactor, as indicated by the arrows 10 and 11.

Disposed upstream of the first reactor plate 1 is a further plate 2 of open-pore ceramic foam, which is not coated with catalyst material. The plate 2 serves

20 as a filter plate for particles contained in the flow of exhaust gas. Furthermore, arranged downstream of the last reactor plate 1 is a very fine filter plate 3 of open-pore ceramic foam without a catalyst coating thereon. The plate 3 has very fine pores and filters

25 out of the gas flow particles of the ceramic base material which have come loose. The spacings between the individual plates 1, 2 and 3 provide for a turbulent effect therein. Those spacings are selected to be non-uniform in such a way that the reaction is

30 as complete as possible, the flow resistance is at a low level and the dimensions of the arrangement are

not excessive. Figure 2 is a diagrammatic view of a two-stage reactor. The two stages are arranged in separate 35 casings or chambers 4 and 5 which are in communication by way of a pipe 6. The waste or exhaust gas to be treated flows into the casing 4 in the direction indicated by the arrow 10. Arranged first in the casing 4 is an uncoated filter plate 2 which 40 is followed at a spacing by two spaced-apart reactor plates 1 which are coated with catalytically active material. In the plates 1, the nitrogen oxides (NO_x) in the gas react with the carbon monoxide (CO) which is also present therein, with the nitrogen oxides being 45 reduced to non-toxic nitrogen. In the pipe 6, air is then added to the exhaust gas by means of a nozzle 7 from a pipe 8, for example by means of a pump in the form of a blower. If it is assumed that the denoxed gas in the pipe 6 still has a component of about 1% 50 CO, then about 0.5% O₂ from the air must be added for complete combustion. With an oxygen component for about 20%, that means the addition of about 2.5% air. As operation is usually conducted

about 5% air.
In the casing 5, the gas again first flows through a filter 2 and then two reactor plates 1, at the surface of which the carbon monoxide undergoes combustion to form carbon dioxide. A very fine filter plate 2 at the outlet of the casing 5 eliminates reaction particles and fine dusts.

at higher than stoichiometry, about double the

55 amount is added in a practical situation, that is to say

One embodiment of the reactor according to the invention used chromium trioxide as the metal 65 oxide. The ceramic foam carrier material was

impregnated with ammonium dichromate solution (between 3 and 25%) and then the substance was thermally decomposed to form chromium trioxide (Cr_2O_3), on the carrier material. The ammonium

70 dichromate may also be reduced with a reducing agent such as for example ethanol or ascorbic acid, to form trivalent chromium trioxide. Fixing on the carrier material is then effected by drying and calcining. The thermal expenditure involved in such 75 a procedure is slight.

Various forms of process may be employed for producing a reactor as described above; for example, when using a slurry of ceramic material to fill an open-pore plastics foam which is then

- 80 vaporised at elevated temperature to produce a porous ceramic foam, the catalyst material such as the non-noble metal oxide referred to above may be added to the slurry used to fill the plastics foam which is then removed in the manner stated.
- 85 Alternatively, the foam may be coated with ceramic material and then impregnated with a solution of a salt of a non-noble metal, the salt then being converted into the metal oxide and fixed on the ceramic foam.
- 90 It will be appreciated that the above-described embodiments of the reactor and process according to the invention have been set forth solely by way of example thereof and that various modifications and alterations may be made therein without thereby
 95 departing from the scope of the invention as defined by the appended claims.

CLAIMS

- An exhaust gas reactor including a refractory carrier means of open-pore ceramic foam, comprising at least two plates which are disposed in succession in the direction of gas flow through the reactor, and a catalytically active surface layer on the
 carrier means.
 - 2. A reactor according to claim 1 wherein the porosity of the plates is between 10 and 60 ppi.
 - 3. A reactor according to claim 2 wherein the porosity of the plates is between 10 and 30 ppi.
- A reactor according to claim 1 and comprising plates of a different configuration or nature.
 - 5. A reactor according to claim 4 wherein the porosity of the plates is of different sizes.
- A reactor according to claim 4 or claim 5
 wherein the plates have different catalytically active surface layers.
 - 7. A reactor according to any one of claims 4, 5 and 6 wherein the plates are of different shapes.
- A reactor according to any one of the
 preceding claims and comprising one or more uncoated plates disposed upstream and/or between and/or downstream of the catalytically coated plates.
- 9. A reactor according to one of the preceding claims wherein the plates are arranged at a spacing
 125 from each other.
 - 10. A reactor according to claim 9 wherein the spacing between the plates varies.
- 11. A reactor according to one of the preceding claims wherein said plates are individually130 interchangeable.

- 12. A reactor according to one of the preceding claims wherein the cataylst material used includes at least one non-noble metal oxide.
- A reactor according to one of the preceding
 claims wherein said catalytically active surface layer is arranged on a base layer (washcoat) of aluminium oxide (Al₂O₃) in the γ-modification form.
- 14. A reactor according to claim 12 or 13 wherein, for a reducing reaction, the catalyst material
 10 comprises at least one oxide of at least one of the following metals: iron, cobalt, nickel, titanium, vanadium, chromium, molybdenum, tungsten, and manganese or a mixture of at least two thereof.
- 15. A reactor according to claim 12 or claim 13
 15 wherein, for an oxidising reaction, the catalyst material comprises at least one oxide of at least one of the following metals; copper, chromium, molybdenum, tungsten, manganese, vanadium, iron or a mixture of at least two thereof.
- 20 16. A reactor according to one of the preceding claims which comprises at least first and second stages, of such a nature that in operation a reducing reaction takes place in the first stage and an oxidising reaction takes plate in the second stage.
- 25 17. A reactor according to claim 16 wherein said first and second stages each comprise at least one plate and they are arranged in a common housing.
- 18. A reactor according to claim 16 or claim 17 and further including an uncoated filter plate
 30 arranged upstream of the first stage in the direction of flow therethrough.
- A reactor according to one of claims 16 to 18 and further comprising an uncoated fine-pore filter plate arranged downstream of the second stage in 35 the direction of flow therethrough.
 - 20. A reactor according to one of claims 16 to 19 wherein air is injected into the flow of gas through the reactor, between said first and second stages.
- An exhaust gas reactor substantially as
 hereinbefore described with reference to Figure 1 of the accompanying drawings.
 - 22. An exhaust gas reactor substantially as hereinbefore described with reference to Figure 2 of the accompanying drawings.
- 45 23. A process for the production of a reactor according to any one of claims 12 to 20 comprising introducing a slurry of ceramic material into an open-pore plastics foam and then removing the plastics foam, wherein said non-noble metal oxide is
 50 added to the slurry used to produce the ceramic foam from the open-pore plastics foam.
- 24. A process for producing a waste gas reactor according to any one of claims 12 to 22 comprising introducing a slurry of ceramic material into an
 55 open-pore plastics foam and then removing the
- open-pore plastics foam and then removing the plastics foam, wherein said non-noble metal oxide is added to a suspension, and the foam which is coated with ceramic material and which has been pre-dried is coated with the suspension, preferably by a
- 60 dipping process, and then it is subjected to calcination.
 - 25. A process according to claim 24 for producing a reactor according to claim 13 or any claim appendant thereto wherein the material (Al₂O₃)
- 65 of the base layer is additionally mixed with the

- suspension.
- 26. A process for the production of a reactor according to any one of claims 12 to 22 comprising introducing a slurry of ceramic material into an
 70 open-pore plastics foam and then removing the plastics foam, wherein the foam which is coated with ceramic material and optionally a base layer and which is in a calcined or uncalcined condition is impregnated with a solution of a metal salt of a said
 75 non-noble metal and the metal salt is then converted into the metal oxide by chemical reaction and/or calcination, and fixed on the ceramic foam.
- 27. A process for the production of a reactor, according to any one of claims 23 to 26 and
 80 substantially as hereinbefore described.
 - 28. An exhaust gas reactor substantially as herein described with reference to the accompanying drawings.

Printed for Her Majesty's Stationery Office by Croydon Printing Company (UK) Ltd, 8/87, D8991685. Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.